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GB 1429476
GB 1376888
GB 1086872
GB 860443
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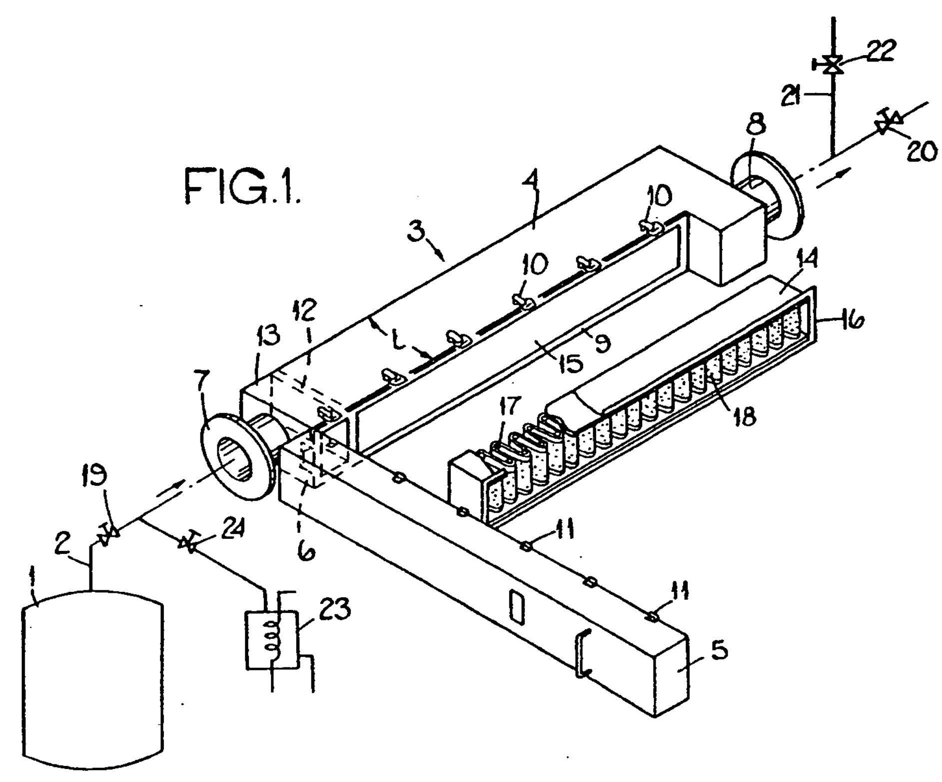
(54) Removing alcohol from waste gas

(57) A layer of activated carbon fibre 18 is disposed in a waste gas flue 2 from an alcohol vapour generating tank 1 so that alcohol is removed from the waste gas passing through the flue 2. The fibre layer 18 may be arranged in a serpentine manner in a frame 14 which may be replacably fitted longitudinally in a housing 4 arranged in the flue, or series layers of particulate active carbon and of the activated carbon fibre may be positioned across a housing arranged in the flue.

FIG.1.



GB 2 022 446 A

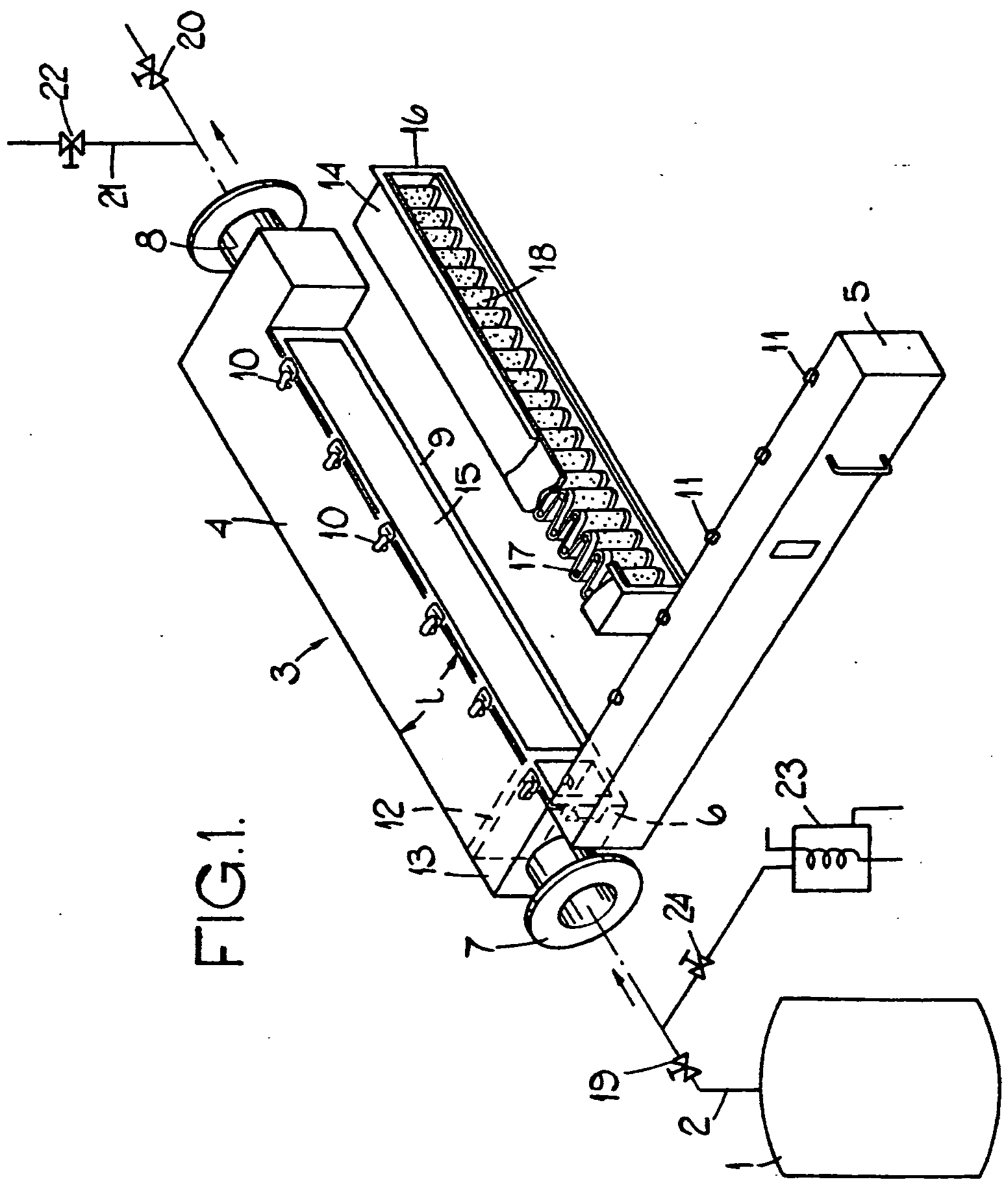
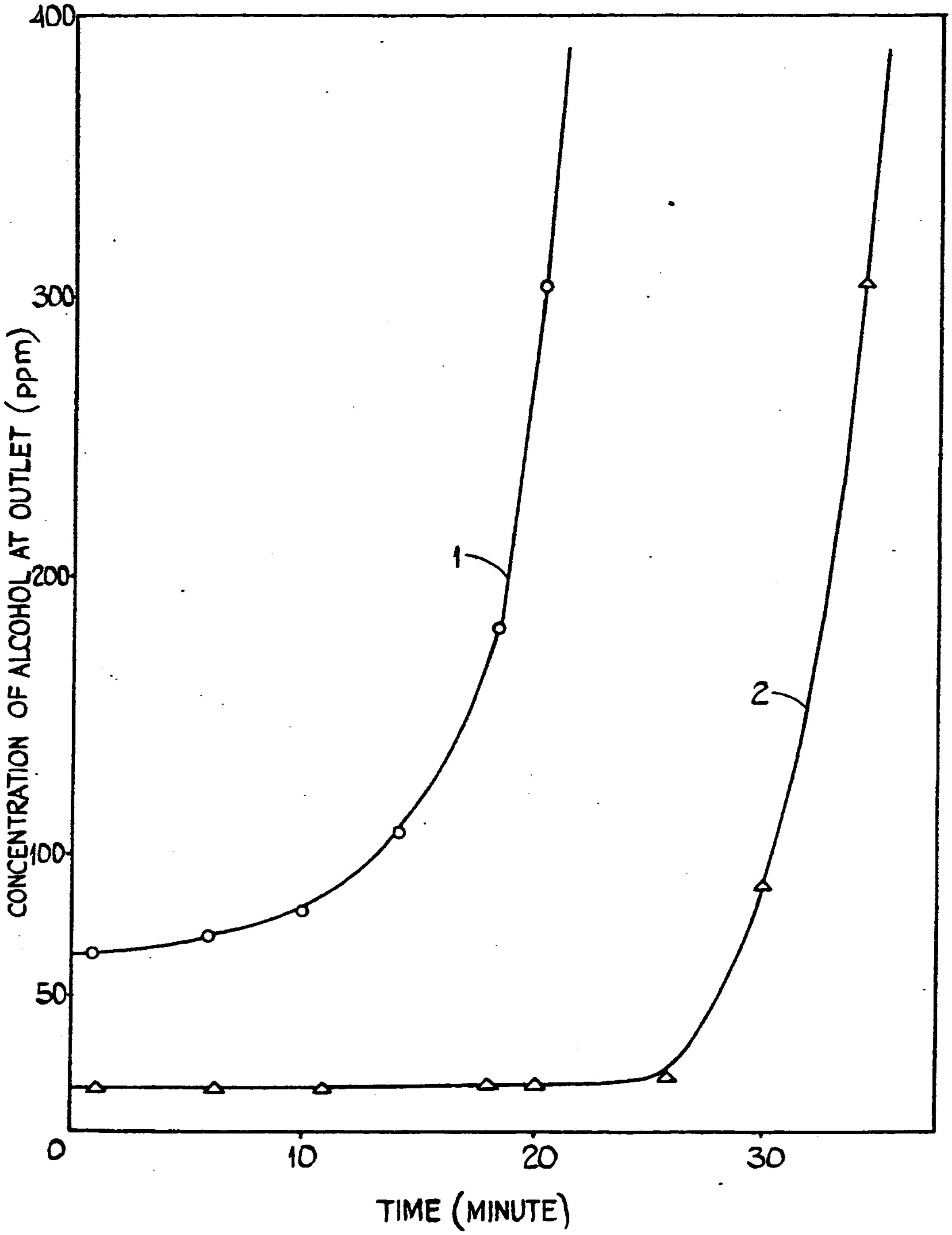


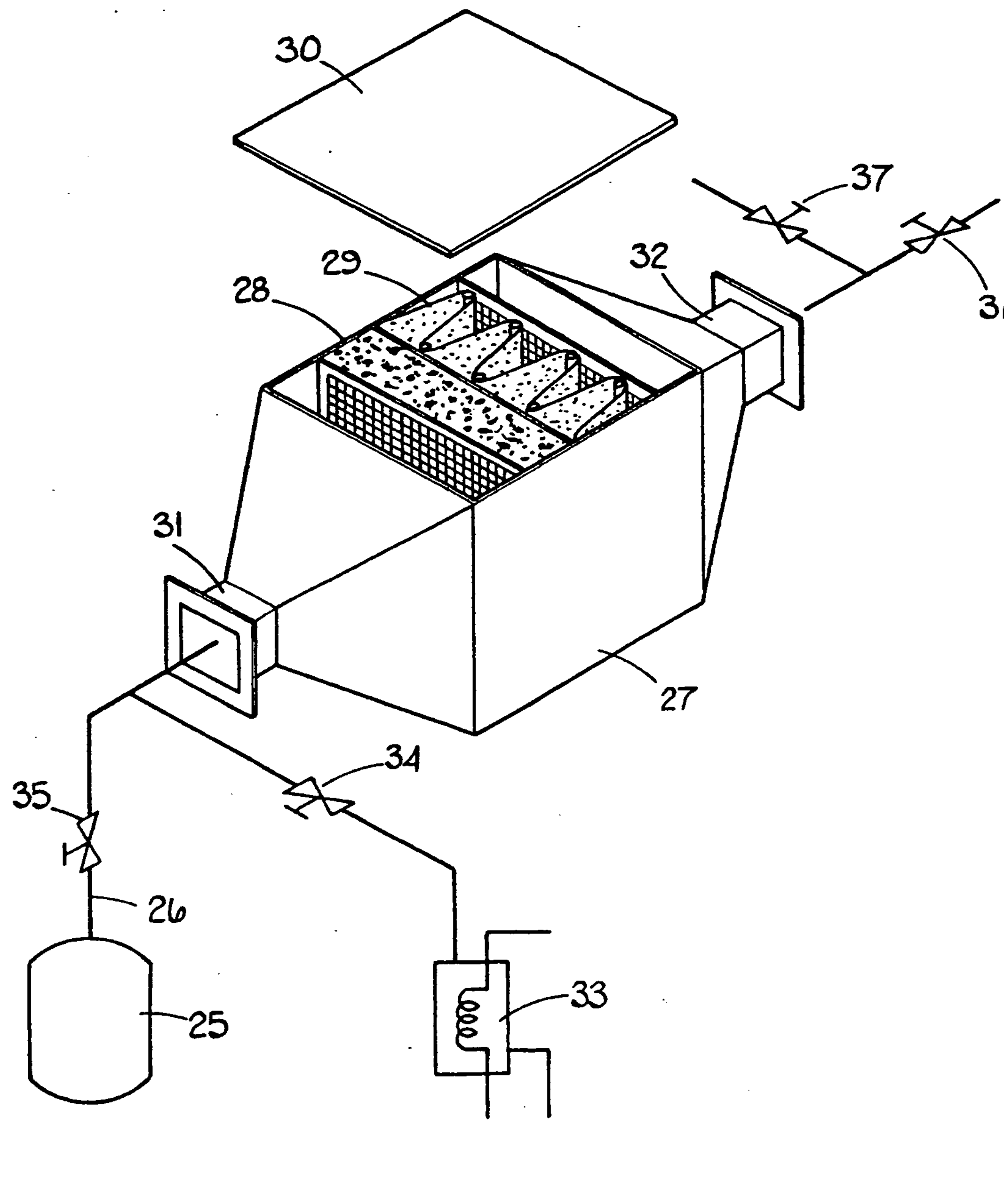
FIG.1.

FIG.2.


400
300
200
100
50
0
10
20
30
CONCENTRATION OF ALCOHOL AT OUTLET (ppm)
TIME (MINUTE)
1
2

2022446

FIG. 3.



SPECIFICATION

A method of and apparatus for controlling alcoholic pollution

This invention relates to a method of and apparatus for controlling alcoholic pollution which occurs in, for example, breweries, distilleries, alcoholic beverage processing plant vinegar manufacturing plants, soy sauce manufacturing plants, printing offices and chemical plants using ethanol.

"Alcoholic pollution" generally refers to pollution believed to be caused by *Pullularia pullulans, Cladosporium herbarum*, and other bacteria that metabolize alcohol and grow in areas where alcohol is manufactured, used or stored. Part of the alcohol manufactured, used or stored in such places evaporates into the air and settles in nearby areas where these bacteria will grow to form a soot-like deposit which blackens or produces black spots on buildings, structures or plants.

Many studies have been made on the prevention of alcoholic pollution and various pollution control methods have been proposed. They consist of simply coating the walls of buildings with a fungi-proof paint, enclosing tanks and other vessels containing alcohol in plants, or in the extreme case, passing any alcohol containing waste gas through a rinsing tower before it is discharged into the air. However, the fact is that alcoholic pollution continues to grow, and some scientists have pessimistically predicted that complete control of alcoholic pollution is impossible based on the current level of technology.

As a result of studies directed towards controlling alcoholic pollution it has now been found that activated carbon fibre is very effective in adsorbing alcohol.

Accordingly, the invention resides in one aspect in a method of controlling alcoholic pollution which comprises disposing a layer of activated carbon fibre along a waste gas flue from an alcohol vapor source so as to remove alcohol from the waste gas passing through said flue.

In a further aspect, the invention resides in apparatus for controlling alcoholic pollution comprising a longitudinal container having a gas inlet and outlet and at least one layer of activated carbon fibre disposed therein.

Any activated carbon fibre can be employed provided the fibre has a sufficient surface area to adsorb alcohol and sufficient mechanical strength to be disposed along a waste gas flue. The surface area preferably is more than about 800 m^2/g, although from the standpoint of mechanical strength the surface area of the fibre is desirably less than about 1500 m^2/g. An activated carbon fibre having a surface area about 900 to 1100 m^2/g is most preferably used. The alcohol adsorption of the fibre having a surface area of about 900 m^2/g is about 200~300 mg per 1 g activated carbon fibre at 20°C.

Suitable activated carbon fibres are, for example, the activated carbon fibres prepared from acrylonitrile polymers or rayon. The activated carbon fibre prepared from an acrylonitrile polymer is most preferred in view of the mechanical strength and alcohol adsorbability of the fibre. For example, a suitable activated carbon fibre can be prepared by heating, at 200 to 300°C under tension, a fibre comprising an polyacrylonitrile or a copolymer containing acrylonitrile as disclosed in Japanese Patent Applications (OPI) Nos. 132193/76 and 137694/76 until the fibre becomes flame retardant, and using superheated steam to activate the resulting carbon fibre until it has a surface area of about 800 to 1500 m^2/g.

The activated carbon fibre can be used in various forms such as a staple fibre, a tow, a woven fabric and a non-woven fabric including felt at a bulk density of about 0.02 to 0.2 g/cc. The thickness of the fibre, tows, and fabrics varies between 10 and 100 mm. Usually it is about 40 to 60 mm.

It is found that an activated carbon fibre layer has far higher ability to adsorb alcohol than other types of adsorbent, or granular active carbon.

The method and apparatus according to this invention for controlling alcoholic pollution is suitable for use in an alcoholic beverage blending tank, a brewery, a distillery, a soy sauce manufacturing plant, a vinegar manufacturing plant, a printing office, a whisky warehouse and any other plant which generates alcohol, as well as tanks and processing apparatus employed in these plants.

The activated carbon fibre is disposed along waste gas flues in a tank or plant in such a manner that it effectively filters the waste gas generated, and it can be disposed in the flue or in an adsorber connected to the flue in any type of structure that provides the largest possible area of contact with the flowing waste gas.

In the accompanying drawings:

Figure 1 is a perspective view illustrating apparatus according to one example of the invention;

Figure 2 is a graph showing a comparison of the ability to adsorb alcohol for two types of active carbon used in Test Example 2.

Figure 3 is a perspective view illustrating apparatus according to a further example of the

invention which combines granular active carbon and activated carbon fibre.

Referring to Fig. 1, in the apparatus of said one example alcohol evaporates from an alcohol beverage blending tank 1, such as a whisky blending tank, as a result of stirring during blending and escapes into the environment together with air when the blended whisky is transferred to and from the tank 1 or when tank 1 is cleaned. A pipe 2 is therefore provided in the top of tank 1 as a waste gas flue and is connected to an adsorber 3. The adsorber 3 comprises an elongated box 4 and a door 5, the two members being connected to each other by a hinge 6 which allows the door to be opened and closed. A waste gas inlet 7 is provided at one end of the elongated box 4 and a clean air outlet 8 is provided at the other end. That portion of the box 4 which abuts the door 5 is provided with a rubber gasket or other type of packing 9 as well as fasteners 10 which, together with hooks 11 provided on the door 5, can bring the box 4 into hermetic contact with the door 5 so that no gas in the adsorber 3 flows out of the box.

Adjacent the waste gas inlet 7 is provided a partition 12 which permits a waste gas inflow to pass through a discharge compartment 13 and enter the space within the door 5. The adsorber 3 also includes a support frame 14 for the activated carbon fibre which is enclosed on all but two longitudinal sides. The frame has a longitudinal dimension that permits its insertion into box 4 through doorway 15 when door 5 is open, but its width is smaller than the depth λ of the box 4. One open side of the support frame 14 is provided with a fixed flange 16 which is of the same dimensions as the packing 9 so that when support frame 14 is inserted into box 4 it is stopped when it reaches a predetermined box depth. A space is then defined between the deepest part of the box 4 and the innermost side of the support frame 14.

The frame 14 is provided with a plurality of supporting rods 17 and a belt of activated carbon fibre 18 is arranged to run in a serpentine manner over the rods 17 from one end of the frame to the other so as to form bellows. The carbon fibre belt is also disposed such that it contacts the inner surfaces of both the top and bottom of the frame. In the example shown, the activated carbon fibre 18 comprises an air permeable felt of the material defined hereinbefore.

In operation, the support frame 14 is inserted into the box 4 until it abuts against the flange 16, whereupon the door 5 is closed to completely isolate the adsorber 3 from the environment. After completion of this preliminary procedure, a valve 19 of the waste gas pipe 2 is opened to allow passage of the waste gas from the tank 1. The waste gas passes through the inlet 7 and discharge compartment 13 and flows into the space within the closed door 5 where it contacts the activated carbon fibre 18. The gas then passes through the fibre 18 and is filtered as its alcohol is adsorbed on the fibre so that on reaching the farthest end of the door 5 clean air is discharged through the outlet 8. The discharged air no longer smells of alcohol or odours characteristic of whisky.

An activated carbon fibre 18 which loses its ability to adsorb alcohol with prolonged use can be replaced with a fresh one by opening the door 5 and removing the support frame 14. The fresh activated carbon fibre 18 is then used to eliminate alcohol from the waste gas by repeating the procedure described before.

Instead of replacing the carbon fibre with a fresh one, the old activated carbon fibre 18 may be regenerated as follows:

A valve 20 at the clean air outlet 8 and a valve 19 on the waste gas pipe 19 are closed, whereas a valve 22 on a heated air branch 21 and a valve 24 on the line extending from a condenser 23 are opened, so that air, inert gas, or superheated steam at 100°C or more is fed through the open valve 22 to heat the activated carbon fibre 18 from which the alcohol is desorbed. At the same time, the desorbed alcohol is transferred into condenser 23 by way of the valve 24, where it is cooled and recovered. The activated carbon fibre from which the alcohol has been removed is subsequently used to eliminate alcohol from the waste gas by repeating the procedure descibed before.

As will be appreciated from the preceding paragraph, the adsorbing action of activated carbon fibre is very temperature dependent. Therefore, to keep the waste gas at a temperature as low as possible, the waste gas pipe 2 is preferably equipped with a cooling device, e.g., a condenser. It is preferable to keep the temperature of the waste gas below 35°C and normally adsorption is carried out at about 20–35°C. It is preferred to keep the temperature as low as possible as long as alcohol contained in the waste gas can be kept in a vapour state. Generally, if the waste gas is at 20°C, 1 g of the activated carbon fibre is capable of adsorbing 200 to 300 mg of alcohol. Accordingly, the type of the material from which the activated carbon fibre is prepared and the conditions under which it is used should be considered to determine how long the activated carbon fibre 18 can be used.

By using the method and apparatus of the above example, substantially every trace of alcohol can be eliminated from the waste gas and hence the possibility of alcoholic pollution is reduced or eliminated.

This invention will now be described in greater detail by reference to the following Test Examples.

Test Example 1

A colony of *Pullularia pullulans* was collected from a blackened wall of a cellar, suspended in sterile water, and cultivated at 20°C for 5 days on a Petri dish containing a Czapeck's medium prepared by the following method.

Preparation of Czapeck's medium

A mixture comprising 30 g sucrose, 2 g $NaNO_3$, 1 g K_2HPO_4, 0.5 g $MgSO_4 7H_2O$, 0.5 g KCl and 0.01 g $FeSO_4$ plus 20 g of agar powder was dissolved in 1 liter of distilled water under heating. The solution was distributed among test tubes until each contained 10 ml of the solution. Each test tube was closed with a cotton stopper and sterilized in an autoclave.

The cultures were then transplanted in a Czapeck's medium and cultivated at 20°C for 5 days to prepare test bacteria.

An agar medium prepared by the following method was poured into the outer groove of each of eight Conway's type diffusion units.

Preparation of agar medium:

2% of agar was dissolved under heating in 0.8% aqueous NaCl solution, and the solution was distributed among test tubes until each contained 10 ml of the solution. Each test tube was closed with a cotton stopper and sterilized in an autoclave.

Subsequently, the eight Conway's type diffusion units were filled with aqueous ethyl alcohol solutions of the concentrations indicated in Table 1, which were so adjusted that the alcohol vaporized at 23°C.

Table 1

Run No.	Concentration of ethyl alcohol solution	Vapour Concentration (23°C)
1	1.27%	250 ppm
2	0.51	100
3	0.13	25
4	0.05	10
5	0.013	2.5
6	0.005	1.0
7	0.0025	0.5
8	0	0

The agar medium in each Conway's type diffusion unit was separately inoculated with the cultured *Pullularia pullulans* test bacteria, and cultivated at 23°C at predetermined ethanol vapour pressures. Table 2 shows the results of cultivation which continued for 12 days.

Table 2

Run No.	Vapour Concentration	Days past 4	6	10	12
1	250 ppm	−	±	+	+ +
2	100	−	±	+	+ +
3	25	−	±	+	+ +
4	10	−	±	±	+
5	2.5	−	±	±	±
6	1	−	±	±	±
7	0.5	−	−	±	±
8	0	−	−	±	±

In the table, − represents "no growth", ± "negligible growth", + "a slight growth", and + + "sufficient growth to produce black spots".

The above table shows that *Pullularia pullulans* grows fairly well at a concentration of alcohol in the range from 25 to 250 ppm, but that it grows only slightly at a concentration lower than 10 ppm.

Test Example 2

Two adsorbers A and B of the structure set forth in Fig. 1 but filled with different types of activated carbon were used to remove alcohol from the waste gas flowing through the waste gas pipe of a whisky blending tank. Adsorber A was filled with a 60 mm thick felt of activated carbon fibre at a felt density of 0.02 to 0.03 g/cc to provide a fibre surface area of 900 to 1000 m^2/g. For adsorber B the support frame of Fig. 1 contained a wire mesh box filled with 60 mm thick particulate active carbon at a density of 0.45 to 0.5 g/cc to provide a fibre surface area of 1000 m^2/g.

The two adsorbers were connected to the waste gas pipe through which a waste gas containing alcohol at 36,400 to 39,300 ppm was flown at a linear velocity of 5 cm/sec. at 20°C and at a relative humidity of 60%, and the change with time in the alcohol concentration at the outlet of each adsorber was measured. Fig. 2 shows the results of the measurement.

In Fig. 2, curve 1 shows the concentration of alcohol at the outlet of the adsorber when granular active carbon is used and curve 2 shows the concentration when activated carbon fibre is used.

As can be seen from Fig. 2, the concentration of alcohol in the waste gas treated by the granular active carbon remained at 60 ppm or higher from the very beginning of the treatment, and was therefore found ineffective in controlling of alcoholic pollution. In contrast with the granular active carbon, the activated carbon fibre was able to hold the concentration of alcohol at a level of 20 ppm or lower for a period of 25 minutes. Therefore, it was found that in actual use, for example, in the case where the alcohol evaporates intermittently the method and apparatus of this invention are capable of effectively controlling alcoholic pollution by replacing the activated carbon fibre every 25 hours.

While the foregoing discussion has only considered the use of activated carbon fibre alone as an adsorbent, it is to be appreciated that activated carbon fibre can also be used in combination with granular active carbon. A granular active carbon used alone is unable to reduce the concentration of alcohol to 60 ppm and below, as demonstrated above. To increase its ability to adsorb alcohol by increasing the thickness of the active carbon layer with which the adsorber is filled is not practical because of the high pressure loss which results as the waste gas passes through the active carbon. However, combining the granular active carbon for the purpose of adsorbing alcohol at concentrations of 60 ppm or higher with an activated carbon fibre for the purpose of adsorbing lower concentrations of alcohol has been found to provide advantageous results in technical and economic terms. Thus, referring to Fig. 3, in the further example shown therein waste gas generated in a tank 25 is fed through a waste gas pipe 26 to an adsorber 27 which is filled with closely arranged combination of layers of granular active carbon 28 and activated carbon fibre 29 and equipped with a door 30, a waste gas inlet 31 and a clean air outlet 32. When this type of adsorber 27 is connected to the waste gas pipe 26 through which waste gas containing 36,400 to 39,300 ppm of alcohol at the inlet is flowing at a linear velocity of 5 cm/sec. at 20°C and at a relative humidity of 60%, the concentration of alcohol measured at the outlet of the adsorber is 20 ppm or less.

Where the adsorbent material is a combination of active carbon fibre and granulated active carbon, it is preferred to use granular activated carbon having a surface area more than about 900 m^2/g. It is desirable to use granular active carbon having as high a surface area possible but usually the surface area does not exceed about 1,200 m^2/g. Usually granular activated carbon having a surface area of about 1,000–1,100 m^2/g is used. The granular size may be about 2 to 12 mesh, preferably about 4 to 6 mesh. The most preferable example of the granular activated carbon is activated carbon prepared from coconut shell. The thickness of the layer of the granular activated carbon may be the same as disclosed above for the activated carbon fibre, i.e., 10 to 100 mm and preferably 40 to 60 mm (of course, the thickness of the granular carbon layer and the activated carbon fibre layer need not be the same). In one practical embodiment of the example shown in Fig. 3, the layers of granular active carbon and activated carbon fibre were 60 mm and 12 mm thick, respectively.

The gas flow rate in the methods described above may be about 3 to 40 cm/sec., preferably about 3 to 15 cm/sec.

CLAIMS

1. A method of controlling alcoholic pollution which comprises disposing a layer of activated carbon fibre along a waste gas flue from an alcohol vapour source so as to remove alcohol from the waste gas passing through said flue.

2. A method as claimed in Claim 1, wherein a layer of granular active carbon is disposed with said layer of activated carbon fibre such that the alcohol-containing vapour passes first through said layer of granular carbon and then through said layer of activated carbon fibre.

3. A method as claimed in Claim 1, wherein after removing alcohol from said waste gas, said activated carbon fibre is regenerated and the alcohol adsorbed on said activated carbon fibre is recovered.

4. A method as claimed in any preceding Claim, wherein said activated carbon fibre is a fibre prepared from an acrylonitrile polymer.

5. A method as claimed in any preceding Claim, wherein said activated carbon fibre is used in the form of a staple fibre, a tow, a woven fabric or a non-woven fabric.

6. A method as claimed in any preceding Claim, wherein said activated carbon fibre is used in a bulk density of about 0.2 to 0.02 g/cc.

7. A method as claimed in any preceding Claim, wherein said activated carbon fibre has a surface area of about 80–1500 m^2/g.

8. A method as claimed in any preceding Claim, wherein the temperature of said waste gas is about −10 to 35°C.

9. A method as claimed in any preceding Claim, wherein said carbon fibre is able to adsorb alcohol at 20°C in an amount of about 200 to 300 mg of alcohol per gram of activated carbon fibre.

10. A method as claimed in any preceding Claim, wherein said alcohol source is an alcoholic beverage blending tank.

11. A method as claimed in any preceding Claim, wherein said alcohol source is a brewery, distillery, soy sauce manufacturing plant, whisky warehouse, or a printing office.

12. Apparatus for controlling alcoholic pollution comprising a longitudinal container having a gas inlet and outlet and at least one layer of activated carbon fibre disposed therein.

13. Apparatus as claimed in Claim 12, wherein a layer of granular active carbon is disposed in said apparatus with said layer of activated carbon fibre.

14. Apparatus as claimed in Claim 13, wherein said carbon fibre and said active carbon particulate are disposed within said container such that said waste gas is passed through said granular active carbon before passing through said activated carbon fibre.

15. Apparatus as claimed in any one of Claims 12 to 14, wherein said activated carbon fibre is prepared from an acrylonitrile polymer fibre.

16. Apparatus as claimed in any one of Claims 12 to 15, wherein said fibre is in the form of a stable fibre, a tow, a woven fabric, or a non-woven fabric.

17. Apparatus as claimed in any one of Claims 12 to 16, wherein said fibre has a bulk density of about 0.2 to 0.02 g/cc.

18. Apparatus as claimed in any one of Claims 12 to 17, wherein said fibre has a surface area of about 800–1500 m^2/g.

19. Apparatus as claimed in any one of Claims 12 to 18, wherein said fibre is capable of adsorbing alcohol in an amount of about 200 to 300 mg alcohol per 1 g fibre at 20°C.

20. Apparatus as claimed in any one of Claims 12 to 19, wherein said fibre is longitudinally disposed in said container in a serpentine manner.

21. Apparatus for controlling alcoholic pollution comprising the combination and arrangement of parts substantially as hereinbefore described with reference to, and as shown in, Fig. 1 or Fig. 3.

22. A method of controlling alcoholic pollution substantially as hereinbefore described.

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